Reference (11)

DECLARATION

I, Maho KASEKI, c/o the Inoue & Associates of 3rd Floor, Akasaka Habitation Building, 3-5, Akasaka 1-chome, Minato-ku, Tokyo, Japan do solemnly and sincerely declare that I am conversant with the Japanese and English languages and that I have executed with the best of my ability this partial translation into English of Unexamined Japanese Patent Application Laid-Open Specification No. Hei 7-179750 and believe that the translation is true and correct.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

December 2nd 2004
(Date)

Maho KASEKI

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(54) [Title of the Invention] Carbon black-containing polyamide resin molded article

(2) At page 2, column 1, line 1 to page 3, column 3, line 39:

[Scope of Claims for Patent]

[Claim 1] A polyamide resin molded article comprising 100 parts by weight of a polyamide resin and 0.05 to 5 parts by weight of carbon black, said carbon black having a hydrogen ion concentration index of 3 or less and a volatile matter content of 10 % or more.

[Detailed Description of the Invention]

[0001]

[Application Field in Industry] The present invention relates to a carbon black-containing polyamide resin molded article. More particularly, the present invention is concerned with a black molded article which exhibits excellent elongation.

[0002]

[Prior Art] With respect to a polyamide resin which is used to produce a molded article, it is a well-known technique to add a carbon black to the polyamide resin for purposes of imparting weatherability to the molded article, coloring the molded article, etc. These purposes can be attained by the

addition of a carbon black to a polyamide resin; however, the resultant carbon black-containing molded article has a problem in that the elongation of the carbon black-containing molded article becomes markedly poor as compared to that of a polyamide resin molded article containing no carbon black and, hence, a breakage or cracking of the carbon black-containing molded article is likely to occur during the use thereof depending on the shape of the molded article.

[0003] In order to solve this problem, various proposals have been made. For example, there have been a proposal to use a specific dye in combination with carbon black (see, for example, Examined Japanese Patent Application Publication No. Sho 60-43379) and a proposal to use a higher fatty amide as a dispersant (see Unexamined Japanese Patent Application Laid-Open Specification No. Sho 61-55146).

[0004]

[Problems to be Solved by the Invention] The improvement in the elongation of the molded article can be achieved by the proposal of Examined Japanese Patent Application Publication No. Sho 60-43379; however, the use of the dye in this proposal causes not only a problem that a molded article obtained by the proposal of this patent document exhibits poor weatherability, but also a problem that, depending on the at-

mosphere in which the molded article is used, the molded article is hazed on the surface thereof or discolors another molded article when these molded articles get in contact with each other.

[0005] On the other hand, in the case of the proposal of Unexamined Japanese Patent Application Laid-Open Specification No. Sho 61-55146, the elongation of the molded article cannot be improved to an expected level. Accordingly, it is an object of the present invention to provide a polyamide resin molded article which exhibits excellent elongation, without using a dye which causes disadvantages in that the weatherability of the polyamide resin molded article becomes poor, and that the polyamide resin molded article is hazed on the surface thereof or discolors another molded article when these molded articles get in contact with each other.

[0006]

[Means to Solve the Problem] The present inventors have made extensive and intensive studies with a view toward solving the above-mentioned problems. As a result, it was found that the problems can be solved by a polyamide resin molded article comprising 100 parts by weight of a polyamide resin and 0.05 to 5 parts by weight of carbon black having a hydrogen ion concentration index (hereinafter, referred to as "pH") of

3 or less and a volatile matter content of 10 % or more.

[0007] More specifically, the present inventors have found that a polyamide resin molded article comprising a polyamide resin and carbon black having a pH value of 3 or less and a volatile matter content of 10 % or more, each as measured in accordance with JIS K6221, exhibits excellent elongation which is comparable or superior to that of a polyamide resin molded article containing no carbon black. Examples of polyamide resins used in the present invention include homopolyamides, such as nylon 6, nylon 66 and nylon 46; copolymers and polymer blends comprising any of these homopolyamides; polymer alloys, such as a nylon 6/rubber alloy, a nylon 66/rubber alloy, and a nylon 66/modified PPE alloy; and copolymers or polymer blends comprising a polyamide and an aromatic nylon, such as nylon 6T/6 and nylon 6T/66.

[0008] The present invention is especially effective when nylon 6 and nylon 66, which are especially likely to be influenced by carbon black, are used. These polyamide resins can be used for producing molded articles, such as an injection-molded article and an extrusion-molded article, which have a relative viscosity in the range of from 1.5 to 10 as measured in accordance with JIS K6810. It is preferred to use the above-mentioned polyamide resins for producing an in-

jection-molded article having a relative viscosity of from 2 to 5.

[0009] When a polyamide resin molded article obtained from any of the above-mentioned polyamide resins has a relative viscosity of 1.5 or less, the elongation of the polyamide resin per se becomes poor and, hence, the resultant polyamide resin molded article is not suitable for practical use. On the other hand, it is difficult to produce a molded article having a relative viscosity of more than 10. Even the production of an injection-molded article having a relative viscosity of more than 5 is difficult. For these reasons, it is preferred that the relative viscosity of a polyamide resin molded article falls within the above-defined range. In the present invention, as mentioned above, the pH value is measured in accordance with JIS K6221.

[0010] In the present invention, the volatile matter content of the carbon black is also measured in accordance with JIS K6221 as in the case of the pH value. The carbon black used in the present invention has a pH value of 3 or less and a volatile matter content of 10 % or more. Preferred examples of carbon blacks used in the present invention include channel black, furnace black and thermal black, which have been chemically oxidized.

[0011] Specific examples of methods for chemically oxidizing carbon black include conventional oxidation treatments using nitric acid or potassium dichromate. It is especially preferred to use channel black which has been oxidized by any of these methods. In the present invention, the term "carbon black content" means the concentration of carbon black in the polyamide resin molded article. Examples of polyamide resin molded articles include a black molded article obtained by a method comprising adding predetermined amounts of the above-mentioned carbon black, a conventional dispersant, a conventional adsorbent, etc. to polyamide resin pellets, melt-kneading the resultant mixture using an extruder to thereby pelletize the mixture, and injection-molding the resultant pellets; and a black molded article obtained by a method comprising adding large amounts of the above-mentioned carbon black, a conventional dispersant, etc. to polyamide resin pellets, melt-kneading the resultant mixture using an extruder to thereby obtain masterbatch pellets, diluting the obtained masterbatch pellets 10- to 100-fold with pellets of a polyamide resin containing no carbon black, followed by mixing, and injection-molding the resultant mixture.

[0012] With respect to the carbon black content of the carbon black-containing molded article, when the carbon black

content is 0.05 part by weight or less, the molded article cannot be colored satisfactorily deep black, and the molded article rather becomes grayish. On the other hand, when the carbon black content of the molded article exceeds 5 parts by weight, the black color of the molded article would not change even if the carbon block content is increased. Thus, the carbon block content outside the above-mentioned range is not favorable from the viewpoint of the desired properties and production cost of the molded article. With respect to the dispersant used in the polyamide resin molded article of the present invention, any conventional dispersant can be used. Examples of dispersants include metal salts of higher fatty acids, such as zinc stearate, magnesium stearate and calcium stearate; and higher fatty amides, such as ethylene bis(stearamide).

[0013] These dispersants can be used individually or in combination, and the amount thereof may be varied in proportion to the amount of the carbon black contained in the polyamide resin molded article. When the dispersant is used in an amount of from 0.02 to 5 parts by weight, relative to 100 parts by weight of the polyamide resin, the dispersant is effective for improving the elongation of the polyamide resin molded article. When the dispersant is used in an amount of

0.02 part by weight or less, the elongation of the polyamide resin molded article cannot be improved to a desired level. On the other hand, when the dispersant is used in an amount of 5 parts by weight or more, the polyamide resin molded article poses a problem in that the operation of melt-kneading of the polyamide resin becomes unstable and, hence, the obtained polyamide resin molded article sometimes fails to exhibit a desired level of elongation.

[0014] In addition to the above-mentioned carbon black and dispersant, the polyamide resin molded article of the present invention may further contain a conventional additive, such as a liquid paraffin, a mineral oil or polyethylene glycol. Further, the polyamide resin molded article may also contain any of other additives, such as a thermal stabilizer and a lubricant; reinforcing materials, such as a glass fiber; and a filler, such as a mineral filler.

[0015] The production of the polyamide resin molded article of the present invention can be performed by a conventional method. For example, the polyamide resin molded article of the present invention can be produced by the following method. An appropriate amount of a mineral oil is blended with polyamide resin pellets in a tumbler or the like. Into the tumbler containing the resultant mixture is charged a mixture of

a predetermined amount of the carbon black used in the present invention and an appropriate amount of the dispersant, and the contents of the tumbler are mixed well, to thereby obtain a homogeneous mixture. Subsequently, the obtained mixture is melt-kneaded using an extruder (e.g., a twin-screw extruder manufactured and sold by Ikegai Ltd.) having a temperature equal to or higher than the melting temperature of the polyamide resin. The thus obtained melt-kneaded mixture is extruded and cut by means of a pelletizer, to thereby obtain pellets.

[0016] The polyamide resin molded article of the present invention can be produced by subjecting the thus obtained pellets to an injection molding. Alternatively, the polyamide resin molded article of the present invention can be produced by a method in which large amounts of the above-mentioned carbon black and additives (e.g., a conventional dispersant) are added to a polyamide resin, followed by melt-kneading using an extruder, to thereby obtain masterbatch pellets, and the obtained masterbatch pellets are diluted 10- to 100-fold with polyamide resin pellets containing no carbon black, followed by injection molding, to thereby obtain a molded article. With respect to the particle size of the carbon black contained in the polyamide resin molded article of the pre-

sent invention, it is preferred that the molded article contains no agglomerated particles of carbon black having a particle size of 20 μm or more, as observed under an optical microscope with respect to a thin section having a thickness of 10 μm, which has been cut out from the molded article.

[0017] It is more preferred that the polyamide resin molded article contains no agglomerated particles of carbon black having a particle size of 5 μm or more, from the viewpoint of the elongation of the molded article. Thus, there is no particular limitation with respect to the method for producing the molded article of the present invention, as long as carbon black particles are dispersed without forming the agglomerates.

(3) At page 3, column 4, line 45 to page 4, column 6, line 2, and Table 1 on page 4:

[0027]

[Examples 13 to 15 and Comparative Example 12] Polyamide resin composition pellets were produced in substantially the same manner as in Example 1, except that nylon 66 (Leona 1300S; trade name) was used as a polyamide resin and carbon black A shown in Table 5 was used. The melt-kneading and ex-

trusion of the polyamide resin composition were performed by using the same extruder as in Example 1. In Examples 13 to 15 and Comparative Example 12, the cylinder temperatures of the extruder were adjusted to 285 °C, 275 °C, 265 °C and 255 °C, respectively. The thus obtained pellets were injection-molded in the same manner as in Example 1, and the resultant molded articles were subjected to a tensile test in the same manner as in Example 1 so as to compare the elongations of the molded articles obtained in Examples 13 to 15 and Comparative Example 12. Further, the sizes of the agglomerated particles of carbon black present in the molded articles were measured.

[0028] The results are shown in Table 5 below. As can be seen from Table 5, the elongation of the molded article obtained in each of Examples 13 to 15 was larger than that of the molded article obtained in Comparative Example 12. Further, the size of the carbon black particles contained in the molded article obtained in each of Examples 13 to 15 was not more than 5 μm.

[0029]

[Table 1]

	Masterbatch						Diluted product			Tensile elongation of the black molded article (in terms of the distance between the chuck grips, %)
	Carbon black			Amount of carbon black	Type and amount of dispersant	Type of polyamide resin	Dilution polymer	Dilution ratio of masterbatch	Amount of carbon black in the molded article	
	Type	pH	Volatile matter							
Ex. 1	A	2.5	22	3 parts by weight	EBS (*) 2 parts by weight	1300S	1300S	20	0.15 part by weight	24.8

Note (*): "EBS" means ethylenebisstearamide and, hereinafter, this abbrebiation "EBS" is used for representing this compound.

(4) At page 5, the bottom line to an upper portion of page 6 (Table 5):

[0033]

[Table 5]

	Amount of carbon black-A (parts by weight)	Type and amount of dispersant (parts by weight)	Cylinder temperature of the extruder (°C)	Elongation of the black molded article (in terms of the distance between the chuck grips; %)	Observation of (large) carbon black particles under an optical microscope (μm)
Ex. 13	0.2	EBS 0.2	285	25.7	Carbon black particles had a particle size of 1 μm or less and were uniformly dispersed in the polyamide resin.
Ex. 14	0.2	- ditto -	275	24.2	- ditto -
Ex. 15	- ditto -	- ditto -	265	22.5	There were some agglomerated particles having a particle size of from 2 to 5 μm.
Compara. Ex. 12	- ditto -	- ditto -	255	13.3	There were some agglomerated particles having a particle size of from 20 to 30 μm.